

Abstract

Title: Homogeneous and Heterogeneous Reaction and Transformation of Hg and Trace Metals in Combustion System

Author: Joseph J. Helble

University: University of Connecticut, Department of Chemical Engineering
191 Auditorium Road, Storrs CT 06269-3222

Contact: Phone: 860-486-4602
Fax: 860-486-2959
Email: helble@engr.uconn.edu

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OBJECTIVE

The objective of this program is to develop improved understanding of the transformations of mercury and other selected trace metals during coal combustion. As part of this effort, models will be developed that can be used to predict metal partitioning between vapor and condensed phase species. It is further expected that for the condensed phase species, these models will provide a dynamic means to predict the distribution between competing surface reactions and condensation as a function of post-combustion conditions. This would represent a significant advance over current models that are interpretive (i.e. that indicate whether condensation or surface reaction was dominant based on a fit to experimental data), and would provide a method for improving predictions of trace metal emissions from a broad range of combustion systems.

To meet this objective, the project will consist of several experimental and model development tasks. These include:

- measurement of heterogeneous mercury oxidation reactions on solid surfaces including fly ash and synthetic fly ash surfaces to determine the effect of individual constituents (including carbon) in the ash, in a flame-based flow reactor in the presence of important post-flame radical species;
- determination of gas-solid reaction rates between As, Cd, Sb, and Se and fly ash constituents as a function of trace metal concentration and gas temperature to obtain fundamental parameters (reaction order, activation energy, pre-exponential factor) needed for modeling metal partitioning;
- measurement of selected trace metal reactions in the flow reactor in the presence of contaminants such as NO_x and SO₂ to determine the effects of these constituents on reaction chemistry;
- further development and expansion of a dynamic model that predicts partitioning by calculating competing rates of condensation and surface reaction for each metal, and thus

incorporates boiler temperature profile, fly ash parameters, and gas phase chemistry.

Regulations to reduce the emissions of mercury from coal-fired power plants were recently announced by EPA. Initial efforts will therefore focus on the homogenous and heterogeneous chemistry of mercury, in an effort to provide timely data and models that can be used in the development of control strategies. Efforts in the later stages of the program will focus on additional trace metals present in coal.

ACCOMPLISHMENTS TO DATE

Although funding was initiated in fall 2005, activity began in earnest in December 2005 when a first year Ph.D. student joined the project. Thus far, efforts have focused on choosing synthetic fly ash materials ($\text{Ca}(\text{OH})_2$, $\alpha\text{-Fe}_2\text{O}_3$, $\gamma\text{-Fe}_2\text{O}_3$, montmorillonite, and kaolinite were chosen for initial study), designing a system for fixed bed studies of heterogeneous mercury reaction chemistry based on bottled gas mixtures, developing a conceptual design for the addition of flame product species to the reaction mixture, and procuring supplies needed to assemble the system and initiate the experimental effort.

In the coming months, it is expected that activity will include heat treatment of these minerals to develop synthetic ash particles, characterization of the synthetic fly ash particle surface area and particle size using scanning electron microscopy (SEM) and BET- N_2 adsorption techniques, conducting initial mercury breakthrough experiments using the fixed bed system with bottled gases and different combinations of synthetic fly ash materials, and conducting initial experiments using flame gases.

FUTURE WORK

Work conducted during the second 6-month period of the project is expected to include experiments using aerosolized fly ash for comparison with fixed bed reactor experiments, and extraction of reaction rate constants for mercury oxidation reactions. Longer term plans include modeling of the heterogeneous reactions occurring for mercury on different fly ash constituents, initial fixed bed experiments for other trace metals including As, Cd, Sb, and Se, and extraction of heterogeneous and gas-solid reaction rate parameters for these metals under post-combustion conditions.

PAPERS, PATENTS, PRESENTATIONS, STUDENTS SUPPORTED

There are no publications, patents, or presentations at this time.

This project is supporting Ms. Clara Smith, a full-time Ph.D. student in the Department of Chemical Engineering at the University of Connecticut.